P102

Dielectric properties of $PbMg_{1/3}Nb_{2/3}O_3$ - $PbTiO_3$ and $BaNb_2O_6$-$SrNb_2O_6$ solid solution ceramics sintered from mechanochemically synthesized nanopowders

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Ferroelectric relaxors are widely used as components of various functional materials exhibiting giant dielectric, electrostrictive, pyroelectric, and piezoelectric responses. Recently we have found out that the use of mechanochemical synthesis enables one to modify substantially relaxor properties of ceramics of the disordered perovskites $PbFe_{1/2}Ta_{1/2}O_3$ and $PbMg_{1/3}Nb_{2/3}O_3$ (PMN) [1, 2]. For both substances high-energy mechanical activation of the starting oxides increases the temperature T_m of the dielectric permittivity maximum and reduces dramatically the frequency shift ΔT of T_m. Similar changes of ΔT and T_m were predicted previously, basing on the first-principle calculations, for PMN with an increased degree of a short-range ordering of Mg^{2+} and Nb^{5+} cations [3]. Such explanation is supported by changes in the magnetic phase transition temperature of $PbFe^{3+}{}_{1/2}B^{5+}{}_{1/2}O_3$ powders (B^{5+} – Nb, Ta) obtained by mechanochemical synthesis [4]. In the present study we investigated the effect of mechanochemical synthesis on the dielectric properties of several solid solution ceramics (1-x) $PbMg_{1/3}Nb_{2/3}O_3$ -x $PbTiO_3$ (PMN-xPT) and (1-x) $BaNb_2O_6$ -x $SrNb_2O_6$ (SBN-x). In both solid solution systems two compositions were studied – one exhibiting good relaxor properties (PMN-0.075PT and SBN-0.75) and the other – a very weak relaxor (PMN-0.33PT and SBN-0.50).

It was found out that high-energy mechanical activation has a very different effect on the dielectric properties of PMN-xPT and SBN-x compositions. While for both usual and mechanoactivated (MA) PMN-0.33PT ceramics ΔT is nearly zero, for MA sample of the relaxor PMN-0.075PT composition the value of ΔT reduces by more than two times as compared to usual ceramics. For both PMN-xPT compositions studied the T_m values of MA ceramics differ by about 10 K from those for the samples obtained by usual method (for PMN-0.075 T_m increases while for PMN-0.33PT it decreases). This difference seems to be due to contamination of the MA samples by iron and formation of the PMN-PT-$PbFe_{1/2}Nb_{1/2}O_3$ solid solution.

For both SBN-x compositions studied T_m values of MA ceramics are lower by more than 50 K while ΔT values are more than twice larger than those reported for single crystals [5] and ceramics obtained by usual method [6]. Similar effects, i.e. lowering of T_m and increase of ΔT values were reported for Fe-doped SBN ceramics [6].

Basing on the comparison of the properties of PMN-xPT, SBN-x and PMN-PFN solid solution compositions fabricated by both the usual solid-state synthesis and by high-energy mechanochemical synthesis one may conclude that the changes in T_m in all the ceramics studied, obtained using high-energy mechanochemical synthesis may be ascribed, at least partially, to the formation of the Fe-containing solid solutions. However the main origin of the ΔT changes observed for MA ceramics seems to be not the incorporation of iron from the milling media into the lattice, but rather the effect of high-energy mechanical activation.

This work was supported in part by RFBR (project 17-03-01293_a) and by the Ministry of Education and Science of the Russian Federation (research project 3.1649.2017/4.6).

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